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NORMAL AND STIMULATED RAMAN SPECTROSCOPY

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Raman Gain Measurements in Diamond

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One of the outstanding problems in the understanding of the stimulated Raman process is the observed sudden increase by 10^6 to 10^{10} in Raman emission at threshold. The first observations were reported in liquids, where, also the measured gain was as high as 100 times that predicted from theory. The discontinuity in intensity and the high gain in liquids have been explained qualitatively by the discovery of self-focusing in many liquids. This same behaviour has also been found in H_2 gas (Bret and Denariez[1], 1966; Avizonis[2] et al 1966) in the absence of self-focusing, although more recently, Lallemand, Simova and Bret[3] (1967) have reported a normal behaviour for H_2 gas. Up to the present time there have been no reports on the Raman intensity behaviour of solids. Since it is known that the angular dependence of stimulated radiation in calcite[4] and diamond[5] obeys theory, it seemed worthwhile to check also the intensity dependence.

The first experiments have dealt with the stimulated emission from diamond. The intensity of radiation at the first Stokes wavelength (λ 7653Å) was measured as a function of incident laser intensity with the arrangement shown in Fig. 1. A giant pulse ruby laser was used as the exciting source. The laser beam was incident on a diaphragm and lens and then fell on the diamond crystal (placed in front of the focus). Simultaneous measurements of the incident laser intensity and the Raman intensity were made with calibrated photomultipliers and a dual beam oscilloscope. The diamond crystal was in the form of a plate 2.2 mm thick with

flat and almost parallel surfaces which were highly polished. It should be noted that because of the large refractive index of diamond, n = 2.5, the reflectivity at the air surfaces is 17 percent. The plate, in effect, behaves as a Raman resonator.

At low incident intensity, spontaneous emission was observed and this increased linearly with increasing laser intensity. At a laser intensity of 0.9×10^9 w/cm^2, the Stokes intensity increased sharply and an increase in laser intensity to 1.5×10^9 w/cm^2 produced a factor of about 10^6 increase in the Stokes output. This behaviour is shown in Fig. 2. At higher laser powers, saturation appeared to set in and further increase resulted in damage to the crystal. Self-focusing was not observed in these experiments and would not be expected to occur at these low powers in diamond.

The sharp discontinuity at threshold observed in the present experiments is to be expected, since, as already mentioned, the diamond plate behaves as a resonator (with mirror reflectivities of 17 percent). The gain, g, may be determined using the results of Fig. 2 and the formula

$$I_s/I_{th} = (R^2 \exp [2 \ell g])^m/R$$

Here I_s is the Stokes intensity for laser intensity I_o, I_{th} is the Stokes intensity at threshold ($\sim 4 \times 10^{-3}$w), R is the reflectivity at the diamond surface ($\sim$ 17 percent) ℓ the length of the crystal (= 2.2 mm) and m is the number of double passes the wave makes during amplification. In our experiment the value of m is 5 and is limited by the beam "walk-off" because of a tilt of 10' of the

diamond surfaces. We obtain for g the value 9.5×10^{-3} I_o/cm, with I_o given in MW/cm^2.

This value compares favourably with the theoretical value calculated from the equation

$$g = \frac{8\pi^2 \nu_s \rho I_o \alpha'^2}{\Delta\nu_s c^2 n^3}$$

where ν_s and $\Delta\nu_s$ are the frequency and full width at half intensity of the Stokes radiation, ρ the number of molecules per unit volume, I_o the laser intensity, c the velocity of light in vacuum, n the refractive index and α' the rate of change of polarizability with normal coordinate. The value of α' has recently been determined from the electric-field-induced infrared absorption in diamond by Anastassakis, Iwasa dnd Burstein[6], $\alpha' = 4 \times 10^{-16}$ cm^2 per unit cell. With this value we calculate for the gain, $g = 8 \times 10^{-3}$ I_o. This is in very good agreement with the value determined from the present experiment and suggests that the available theory is applicable to diamond. It is intended to carry out a similar experiment with calcite.

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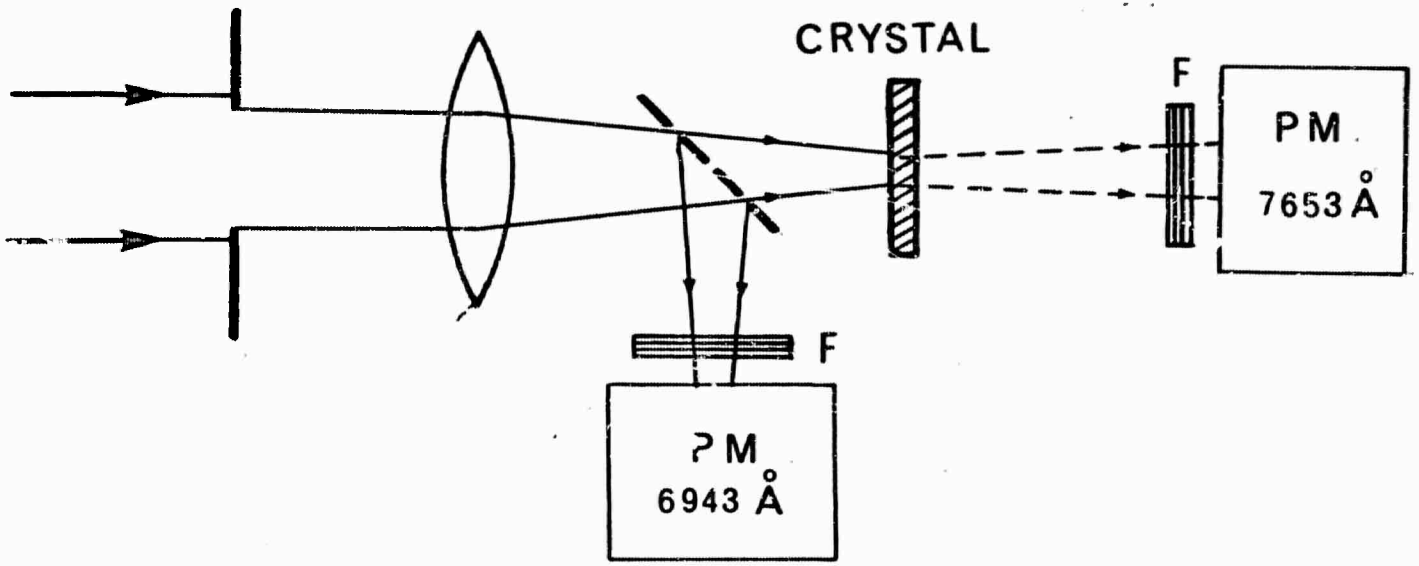

Fig. 1.

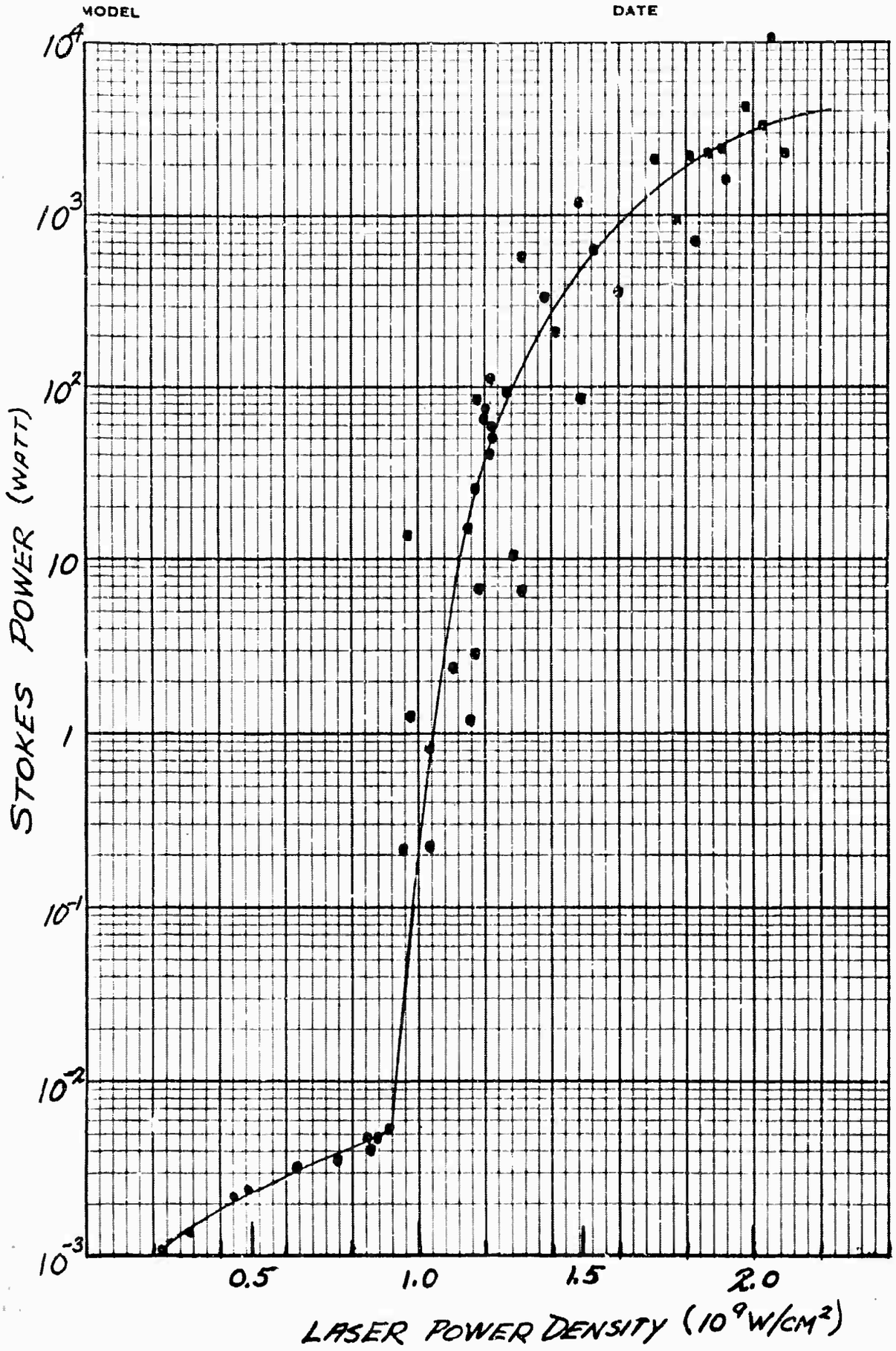
MODEL
DATE
10^4
10^3
10^2
10
1
10^{-1}
10^{-2}
10^{-3}
STOKES POWER (WATT)
0.5
1.0
1.5
2.0
LASER POWER DENSITY (10^9 W/CM^2)

Fig. 2

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Up to the present time measurements of Raman gain have been reported for several liquids and hydrogen gas, but not of solids. We have measured the gain in a diamond crystal. At low laser intensity, spontaneous emission was observed and this increased linearly with increasing laser intensity. At a laser intensity of 0.9×10^9 W/cm^2, the Stokes intensity increased sharply and at slight increase in laser intensity produced an increase of 10^6 in Stokes output. In our experiments the sharp increase at threshold is expected since the diamond crystal was in the form of a plate and behaved as a resonator. The measured gain/cm is 9.5×10^{-3} I_0 where I_0 is the laser intensity given in MW/cm^2. This agrees very well with the value $g = 8 \times 10^{-3}$ I_0, calculated from the data on normal Raman scattering.

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